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Mechanical nanomanipulation of single strain-induced semiconductor quantum dots

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We report on low temperatures (4 K) *in situ* nanomanipulation of the confining potential of single strain-induced $Ga_{0.9}In_{0.1}As$ quantum dots. This was achieved by scanning a metal coated tapered optical fiber tip over the self organized InP stressor islands that are responsible for the localized strain field in the GaInAs/GaAs quantum well. By scanning the tip with a shear force contact of the order of 1 nN, we thinned down the InP stressor islands in an unexpectedly reproducible and controlled way. The modification of the confining potential was directly monitored by measuring *in situ* the photoluminescence of each manipulated dot using a near-field scanning optical microscope.
 [S0003-6951(99)03829-2]

Strain induces modification in the energy band structure of semiconductors. This very principle has been put into application in order to tailor one-[1] and zero-[2] dimensional confining potential for charge carriers and excitons in semiconductors heterostructures. In particular, InP islands grown on GaAs/GaInAs films produce localized strain fields in the immediate proximity of the dots[2] owing to the lattice mismatch between the materials. In this case the InP islands are acting as ''stressors'' while the actual quantum confined potential is located the InGaAs film. The photoluminescence (PL) of such strain-induced quantum dots is very reminiscent of atomic optical spectra[3] making such systems potentially promising candidates for optoelectronic devices. This is particularly attractive since a large ensemble of such dots can be grown on typical wafer size areas using molecular beam epitaxy or metalorganic vapor phase epitaxy deposition in coherent Stransky–Krastanov mode.[4] Despite the enormous progress achieved in sample quality, the PL spectra dots are broadened by inhomogeneities in the size distribution of the confining potential.[5] In the present work we show that the dot potential can be, to some extent, tailored by mechanically removing some of the stressor material using a tuning fork shear force microscope. Figure 1 shows the structure of the sample under investigation. It consists of a single 7-nm-thick $Ga_{0.9}In_{0.1}As$ quantum well imbedded in GaAs barriers material. The InP islands are formed by coherent Stransky–Krastanov growth mode on top of the 5 nm GaAs barrier film.[2] Such InP stressors are typically 22 nm high and have a diameter of 80 nm as analyzed from noncontact scanning force microscopy topography images. The confining and PL emission energies are schematically shown in the inset of Fig. 2. When a single dot is directly illuminated, up to three PL peaks are detected.[6] The $s-s$, $p-p$, and $d-d$ transitions correspond to the electron-hole exciton recombination in the first three confined energy levels. The denomination in terms of *s,p,d* for electron and hole levels is done in analogy with atomic spectroscopy. The basic idea behind this work is illustrated in the right-hand side panel of Fig. 1. The InP islands can be mechanically modified by using a tip that is being scanned in contact with the sample with a controlled shear and friction force. By removing the appropriate amount of InP material the strain field can be diminished in a quasi-monotone and controlled way.

The mechanical manipulation of individual InP dots was performed in a top-loading low-temperature near-field scanning optical microscope (NSOM). The tip serves a dual purpose, that of manipulating the dots as well as to provide for local illumination in order to analyze their PL. Cryogenic temperatures are necessary in order to obtain a strong enough PL signal. The microscope is designed to fit inside a 50.8 mm outside diameter vacuum tight inset so that it can

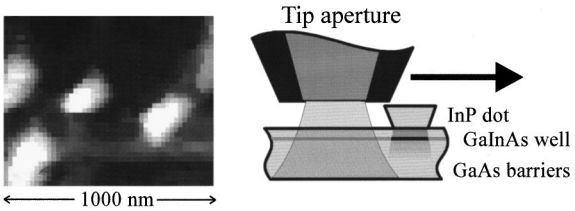

FIG. 1. Left-hand side: shear force topography of InP islands taken at 4 K with a tapered metal coated optical fiber tip, the full contrast represents 20 nm in height. Right-hand side: schematic view of the sample and the optical tip aperture illuminating ($\lambda = 633$ nm) and scanning in shear force engagement over the dot.

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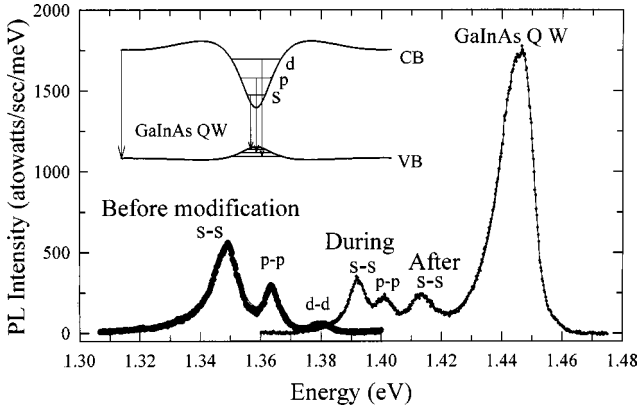

FIG. 2. Inset: schematic band diagram along the plane of the quantum well (GaInAs QW) in the vicinity of an InP stressor dot. The strongest PL peak at 1.445 eV originates from the unstrained regions of the QW. Heavy symbols: PL spectrum of the $s-s$, $p-p$, and $d-d$ transitions in a single dot before manipulation. The peaks at 1.39 eV ($s-s$) and 1.40 eV ($p-p$) originate from a dot being ''milled'' with a shear force of about 1 nN. The peak at 1.41 eV originates from a neighboring dot for which the ''milling'' has just been completed.

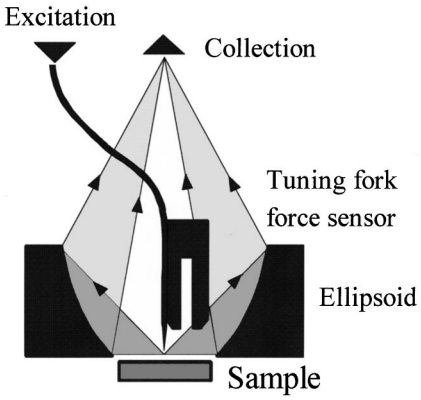

FIG. 3. Schematic representation of the low-temperature near-field scanning optical microscope head.

be plunged in a liquid He bath cryostat. The sample and the microscope are cooled down to 4.2 K in low pressure He exchange gas. PL with high spatial resolution is obtained by bringing the aperture at the apex of an aluminum coated tapered optical fiber tip into a close proximity with the sample surface.[7] The optical probe is used in illumination mode in order to excite the sample surface with the 632.8 nm line of a HeNe laser. We used a probe with a tip aperture size of 160 nm$\pm$10 nm as characterized from far-field angular resolved emission measurements.[8] The reflected signal containing the PL signal is collected over 70% of the total solid angle available in reflection using an ellipsoidal aluminum mirror. The collected PL signal is then dispersed on the grating of a 640 mm focal length spectrometer of numerical aperture 0.1, and detected with a nitrogen cooled Si-chip CCD camera. The spectrometer resolution was set to 0.5 meV. The sample was mounted on a piezoelectric scanner providing a total of 1.4 μm$\times$1.4 μm scan range under 0 to +100 V drive signal. Rough positioning of the sample was achieved over $5\times5\times5$ mm in x, y, and z using a low-temperature low-voltage piezoelectric inertial positioner. The PL signal was integrated over 10 s per tip position pixel (i.e., 14 nm) over the whole measured spectral range. The tip–sample force control is achieved using a piezoelectric tuning fork force sensor[9] allowing to set the tip–sample shear and friction force interaction ranging from about 0.1 pN to several tens of nN. Quantitative details concerning the determination of forces can be found in Ref. 10. A schematic description of the microscope is shown in Fig. 3. In order to modify the InP stressors, the tip–sample interaction is adjusted with forces of the order of 1–2 nN while scanning at a rate of 1.4 nm/s. The PL spectra were monitored during this process. InP being a much more brittle material than the Al coated fiber tip, we were able to preserve the integrity of the tip apertures during the islands ''machining.'' It should be noted however that when shear forces exceed the range of 2–5 nN the aluminum coating of the tip is pealed off and the optical aperture is destroyed. On the other hand shear forces much lower than the nN range do not modify the dots and can be considered noninvasive. The tuning fork holding the tapered optical fiber tip is directly mounted on a specially developed low temperature preamplifier in order to minimize the parasitic capacitive cable losses of the weak piezoelectric shear and friction force signal. Topography of the sample can also be obtained in a noncontact using friction force imaging mode. To do this, the tip aperture peak-to-peak excursion at the resonance frequency of the fork (33 kHz) is adjusted to be 50 pm. The friction force is then kept at 30 pN using a feedback loop on the tip–sample distance. A typical PL spectrum taken in a noninvasive friction tip–sample distance control is shown on the lower left-hand side of Fig. 2. In this mode of reduced friction, no modification of the dots was seen even after 50 consecutive scans over the same dots. We then adjusted the shear force to be of the order on 1 nN and performed two consecutive line passes over what appears to be four individual dots. At such a force level modification of the InP island was achieved. Figure 2 shows one representative PL spectrum out of 100 at a position in the scan for which a first dot has been thinned and a second dot is in process of being modified. A complete PL spectrum is acquired each time the tip traveled 14 nm along its scan path. Figure 4 shows a gray scale intensity map of all the corresponding acquired PL spectra. The lower panel of Fig. 4 shows the result of a first tip pass over the dot. The result of the second tip pass is shown in the middle and upper panels. In both scans, the ground state ($s-s$) excitonic transition [as well as the ($p-p$) when visible] are seen to shift away from its original energy at 1.348 eV toward the PL energy position of the unstrained GaInAs QW at 1.445 eV. This is an irreversible process which shows up in a change in the sample topography. Each passage contributes to further increase the energy of the dotted PL peaks until they disappear in the QW PL peak. After several scans over the same sample area, all the dots appear to have been erased or significantly smeared out from topography images. The edges of the scan area however reveals mounds probably originating from the excess InP material that has been taken away from the dots. Similar results were obtained by imaging the sample under the tip of a commercial atomic force microscopy (AFM) microscope used in contact with 1 nN loading vertical force. While the first topographical image revealed the dots with their expected sizes, subsequent images also showed that the dots were thinned down and laterally smeared out. Mounds

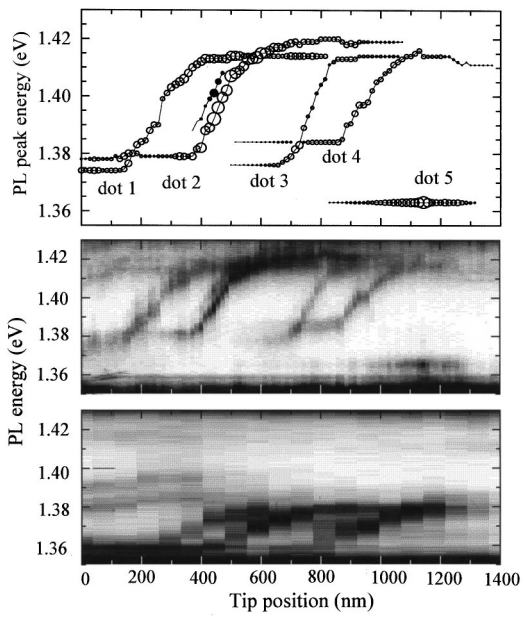

FIG. 4. PL spectra during mechanical modification of dot. Lowest pannels: gray scale rendering of the PL spectra as a function of the tip position along a line scan. White region corresponds to low level PL signal. Dark indicates higher PL intensities. The lowest panel represent the result of a first tip passage which ''pulls'' the dot PL peak out of that of the dot ensemble. The two upper panels show the result for a second passage over the same scan line, revealing that four dots at least are being mechanically modified. For shear forces of the order of 1 nN, the process is irreversible. The size of the symbols in the upper panel is proportional to the PL peak strength.

of materials were also seen at the edges of the scanning range. The amount of material that is taken away after each passage was however, difficult to estimate. The middle and upper panel of Fig. 4 shows in great detail that the process of the dot strain potential modification can be accurately controlled on four individual quantum dots. The scanning length over which a full modification of the dot is done is about 200 nm which is consistent with the physical size of the Al coated fiber tip apex. The fact that the strain potential has been reduced, can be further confirmed from Fig. 2 where it can be seen that the energy separation between the $p-p$ and $s-s$ PL exciton peak is diminished by more than a factor two from its original value. This energy separation is directly related to the strength of the confining potential. A fifth dot can also be seen in Fig. 4. Its PL energy is totally unperturbed by the tip. This is possibly due to the fact that the tip is only passing near the dot without affecting it. A careful inspection of the upper panel of Fig. 4 shows that the PL of the dots is visible up to 450 nm before or after the tip passed over or near the dot. This effect is due to the fact that during their lifetime, the photogenerated excitons can diffuse over a certain length before they recombine. Such a diffusion length can be inferred from the data by plotting the PL intensity as a function of the tip–dot distance. This is shown in Fig. 5. As a guide to the eye we have plotted an exponentially decaying function with a typical length of 150 nm. This length can be seen here as an effective exciton diffusion length. In order to get more quantitative information on the real diffusion

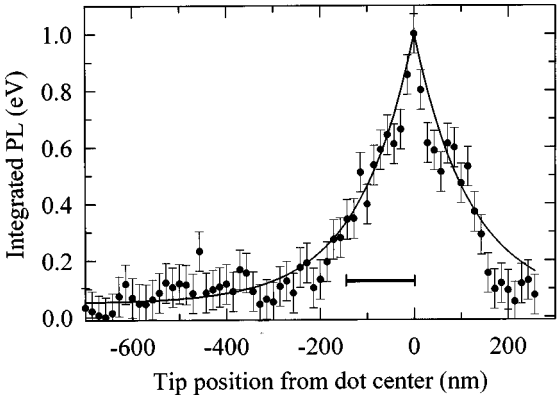

FIG. 5. PL intensity as a function of the tip aperture position passing nearby a single isolated dot (dot 5 of Fig. 4). The full line is an exponential fit to the data. The small horizontal bar indicates the measured total tip aperture diameter 160 nm±10 nm.

length, one would need to solve for a two-dimensional diffusion model the tip illumination beam profile as well as a precise knowledge of the position of the tip in relation to the dot center. Figure 5 shows that the spatial resolution in the mapping of the ground state PL intensity is essentially limited in by the exciton diffusion. The probe diameter aperture (160 nm±10 nm) measured according Ref. 8 is about 3 times smaller than the spatial distribution of the PL intensity and has only a minor contribution to the diffusion limited spatial resolution.

Being in a position to tailor the emission energy of individual quantum dots is helpful in studying their PL line shape. In an ensemble of close-packed nearly identical quantum dots, the PL can originate from several dots within diffusion distance from the tip aperture. In contrast, a modified dot can be singled out from the ensemble. This was done and allowed for more accurate studies of the PL line shape of a single stressor induced dot. The results will be reported elsewhere.

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